

FY05 LDRD Final Report Technology Basis for Fluorescence Imaging in the Nuclear Domain (FIND)

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Abstract

Work performed as a part of this ER sets the foundation for applications of high brightness light sources to important homeland security and nonproliferation problems. Extensive modeling has been performed with the aim to understand the performance of a class of interrogation systems that exploit nuclear resonance fluorescence to detect specific isotopes, of particular importance for national security and industry.

Introduction/Background

We outline work that will set the foundation for applications of high brightness light sources to important homeland security and non-proliferation problems. These applications would exploit nuclear resonance fluorescence (NRF) to enable detection of dangerous materials. NRF-based interrogation technologies are relatively unexplored, qualitatively different, and potentially very powerful. For these reasons we judge that work described here has the potential to become important for several lab missions. Nuclear resonance fluorescence refers to the absorption and reemission of photons by a nucleus. From the perspective of efforts aimed at detecting the composition of clandestine shielded materials this process has two quite useful characteristics. First, the pattern of electromagnetic resonances exhibited by a nucleus serves as a unique fingerprint identifying that isotope. This means NRFbased interrogation systems can unambiguously determine details of the isotopic composition of cargo. The second useful feature of nuclear photo-absorption relates to the sheer strength of absorption resonances. In some cases the cross section for absorption of deeply penetrating high-energy photons is many times larger than cross sections characterizing photoatomic interactions. Such strong resonances have been found in a number of actinides including ²³²Th, ²³⁶U and ²³⁸U. Existence of pronounced resonances allows detection of even very small quantities of these isotopes in large well-shielded containers. No other interrogation technology comes close, e.g. to being able to detect and identify a golf-ball size amount of ²³²Th in a packed car trunk.

One specific example of the promise offered by non-intrusive photon interrogation relates to the pressing national problem of detecting special nuclear material hidden in sea-going cargo containers. This is a daunting challenge because these large containers carry essentially every material found in modern commerce and must be scanned quickly for practical reasons. A technical report [1] studied the efficacy of NRF-based cargo interrogation using next generation photon beams. It was found that this system is characterized by a simple detection protocol, uniquely high efficiency and quantifiably low false positive/negative error rates. Error rates and absolute detection efficiencies of other proposed interrogation technologies have yet to be clearly quantified. Initial comparisons presented in [1] suggest that NRF

interrogation with narrowband sources may provide a markedly valuable tool with which to address "the cargo container problem".

A few efforts are needed before nuclear resonance interrogation technologies can be practically implemented. One of these relates to basic uncertainties about the structure of the interesting fissile nuclides ²³⁵U and ²³⁹Pu. Before the electromagnetic resonances can be exploited to detect these isotopes, the resonances have to be first discovered. Second, more mature modeling work investigating the performance of NRF interrogation systems is needed. Nuclear electromagnetic resonances have very small effective widths – typically on the order of 1 eV. Any photons produced outside of this range are essentially useless and only serve to contribute unwanted background and radiological dose. This is why commonly available bremsstrahlung sources are poorly suited for NRF interrogation.

Research Activities

Though [1] presented careful estimates for the detection power of NRF interrogation systems, some details of photon transport in these systems need more consideration. This will inform future decisions about detector and light-source design, as well as help solidify an understanding of the capabilities of these detection systems. To understand the specific challenges, note that proposed NRF detection systems rely on observing the depletion of resonant photons occurring as an interrogating beam passes through dangerous material. This depleted photon region - or "notch" - can be replenished when photons with energies slightly larger than the resonance energy undergo small angle Compton scattering. Once this occurs the detection system fails to notice clandestine material. Our initial calculations suggest that this problem is severe for bremsstrahlung machines interrogating thick cargos. Monte Carlo and analytic calculations suggest that the notch re-filling is negligible for narrow-band sources like T-REX. [2] Careful definitive work is needed for a clear understanding of when these systems fail.

With a careful Monte Carlo study of photon transport we will be in a position to specify details of a test interrogation system. This involves determining requirements for the light source and segmented photon detector arrays. Complete specification for the design and performance of a test interrogation system would serve as a starting template for future systems.

Results/Technical Outcome

The results of the modeling effort to understand the basic detection limits of NRF systems have been submitted for publication [3] and are summarized here. Fig. 1 gives a schematic outline of the NRF imaging system studied. In this system collimated pulses of photons are sent to interrogate cargo. The light source is tuned so that some of these photons have just the right energy to resonantly excite an electromagnetic transition in the nuclide we want to find.

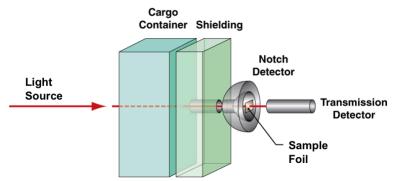


Figure 1. Schematic representation of the detection system studied. A photon beam is sent to interrogation cargo. After passing through the container the flux of resonant and off-resonant photons is measured. Resonant flux is measured by "notch detectors" that observe NRF within a small sample foil made of the isotope that is being looked for. The flux of off-resonant photons is measured with a simple transmission or currect detector.

The basic detection method is loosely analogous to conventional X-ray imaging. Specific isotopes are detected by looking for the shadows they cast in the unscattered beam transmitted through cargo. To illustrate this we show in Fig. 2 the evolution of the photon spectrum of a beam which passes through cargo containing a small amount of uranium. The energy of the beam has been chosen to coincide with a pronounced resonance in this uranium isotope. The top line in this figure corresponds to the spectrum after two optical depths of water. Apart from a small bump arising from NRF emission in the U, this spectrum is essentially flat over relatively large energy ranges (~100 keV). As the beam passes through uranium those photons that resonantly excite nuclei are preferentially absorbed. This results in the prominent depletion of resonant photons, or "notch", that indicates the presence of clandestine material. The two lower curves in Fig. 2 show this notch. If the suspected material were not present in the cargo, the beam exiting the container would not contain a notch. Instead only the smoothly varying attenuation characteristic of Compton scattering and pair production would be evident.

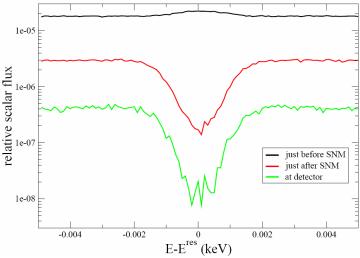


Figure 2. Calculations illustrating the generation and evolution of a "notch" in a narrow beam interrogating cargo. Here the cargo container is assumed to be uniformly filled with water representing a total optical depth of 4 to 2 MeV photons. At the center of the cargo is a disk of uranium with diameter approximately 2 cm. This uranium is assumed to have an M1

resonance at excitation energy $E^{res}=2$ with a width $\Gamma_0=30$ meV similar to those found in well-studied actinides. The line labeled "just before" corresponds to the flux of photons at a point near where the beam enters the uranium. The small bump near the resonance energies occurs because of re-emitted NRF photons that scatter at large angles relative to the incident beam. The line labeled "just after" corresponds to the flux of photons at a point near where the beam exits the uranium. The line labeled "at detector" corresponds to the photons spectrum recorded by an ideal detector positioned outside of the cargo container.

Fig. 5 shows the dependence of the needed number of expected counts on the effective notch refilling fraction α . Results for both static and dynamic scanning are shown. In the static case the expected number of counts is chosen ahead of time. For the dynamic case interrogation is assumed to cease immediately following an observation inconsistent at the six sigma level.

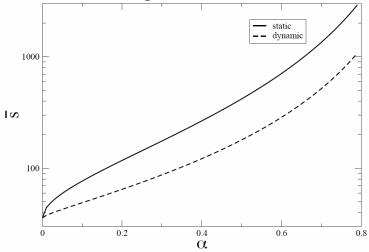


Figure 3. Dependence of the number of expected counts needed to achieve error rates smaller than 10^{-9} on the effective notch refilling fraction α . The line labeled "static" is calculated by assuming that collected signals are analyzed only after a scan has been completed. The line labeled "dynamic" is calculated by assuming that interrogation stops as soon as the observed number of resonant photons gives a definite answer about whether or not the cargo contains clandestine material.

Summary

Many nuclei exhibit electromagnetic resonances that can serve as reliable and readily observable signatures of even small quantities of material. Here we have studied one detection method that can be viewed as the nuclear analog of conventional X-ray imaging. Every indication is that these systems will work and will be characterized by high scanning efficiency and quantifiably small error rates. Well studied actinides, for example, have resonances that are estimated to enable detection of even a small amount (linear dimension ~ 1 cm) of these materials hidden in a cargo representing 10-15 optical depths to MeV protons. This corresponds to a rather large areal density of order 300 g/cm². No other interrogation system boasts the capability to accurately detect small amounts of particular isotopes in such thick systems.

The efficacy and performance of different light sources for NRF-based detection has also been considered. Broadband bremsstrahlung sources can be used to detect material in thick systems. However, the radiological dose and scan time per unit integrated brightness are large. High-brightness sources in which gamma-rays are generated by scattering laser light from energetic electrons are more naturally suited to NRF-based detection. These sources are associated with a radiological dose and

scan time per unit integrated brightness that is about one thousand times smaller than for bremsstrahlung sources.

We have also studied a possible failure mode of these systems related to small angle Compton scattering and the resulting "notch refilling" that erases the signature of clandestine material. Simulations and analytic estimates show that notch refilling is not important for light sources with fine energy and angular resolution. For the spectral and angular resolution characterizing photon beams produced by Compton upscattering of laser-light, the notch refilling fraction is estimated to be less than 1% even for a cargo with an optical depth of 100. For bremsstrahlung sources notch refilling is more serious, but still not pronounced as long careful beam and detection collimation is employed.

References

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 [2] Barty, C.P.J. & Hartemann, F.V., "T-REX: Thomson-Radiated Extreme X-rays", UCRL-TR-206825 (2004).
- [3] Pruet, J., et al., "Detecting Clandestine Material with Nuclear Resonance Fluorescence," submitted to J. Appl. Phys. (2005).